Supporting information

Evidence for the coexistence of polysulfide and conversion reactions in the lithium storage mechanism of MoS_2 anode material

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Figure S1. (a) SEM image and (b) EDS mapping spectrum of commercial MoS_2.

Figure S2. Low-magnitude TEM images of a MoS_2 microsphere for (a,b) pristine, (d,e) fully discharged, and (g,h) fully charged MoS_2. Corresponding ED patterns of the MoS_2 electrode in (c,f,i), respectively.

Figure S3. Voltage profile (left, gray line), and Mo valence vs. capacity (right, dark blue line) of MoS_2 electrode upon the first cycle. Mo valence was calculated via linear interpolation using the X-ray absorption edge point of bulk MoS_2, and Mo metal foil as the reference.

Figure S4. Voltage profiles for MoS_2 and MoS_2+25S composite during the first cycle at 100 mA g^{-1} between 3.0 and 0.001 V (*vs.* Li/Li^+). The highlighted region indicates that the additional part which is only related to the sulfur reaction.

Figure S5. Voltage profile (left, gray line), and Mo valence vs. capacity (right, dark blue line) of MoS_2 electrode upon the second cycle. Mo valence was calculated via linear interpolation using the X-ray absorption edge point of bulk MoS_2, and Mo metal foil as the reference.

Figure S6. Depth profile XPS of MoS_2, which was obtained before sputtering and after sputtering at 30 nm (5 nm per each level). (a) Mo_{3d} XPS spectra of 1FC and 2D200, (b) S_{2p} XPS spectra of 1FC and 2D200.

Figure S7. XPS spectra of MoS_2 after eliminating the SEI layer. (a) Mo_{3d} at level 6 (30 nm) and (b) S_{2p} at level 6 (30 nm).

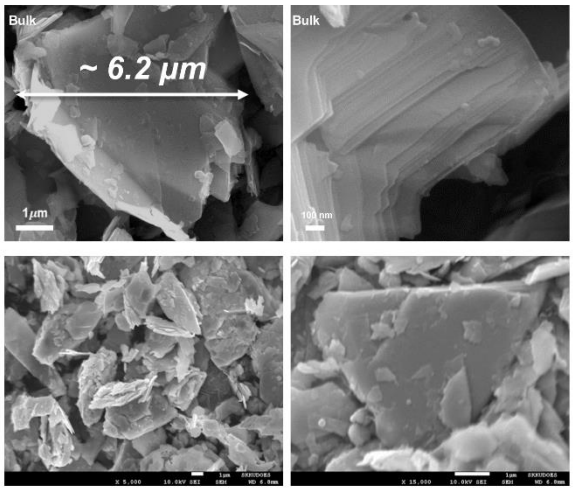
Figure S8. k^3-weighted Fourier transform magnitude and imaginary part along with the best-fit model of Mo K-edge EXAFS spectrum of pristine MoS_2, 1st full charge, and 2nd discharge 400.

Figure S9. k-weight fitting of pristine MoS_2 EXAFS to determine the reduction factor (S_0^2) and coordination number (CN).

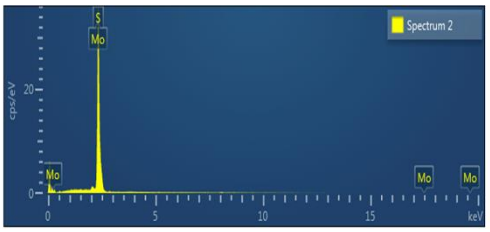
Figure S10. (a) Voltage profiles of the MoS_2 during the 1st to 10th cycles between 0.001–3.0 V (*vs.* Li/Li^+) at a current density of 100 mA g^{-1}. (b) CV profiles between 0.001–3.0 V (*vs.* Li/Li^+) at a scan rate of 0.05 mV s^{-1} during the 1st to 10th cycles.

Table S1. Best-fit results for k^3-weighted Mo K-edge EXAFS spectrum data.

(a)



(b)



Element	wt %	atomic %	standard level
Mo	60.28	33.65	Mo
S	39.72	66.35	FeS_2
Si	0.00	0.00	SiO_2
N	0.00	0.00	BN
P	0.00	0.00	GaP
total	100	100	

Figure S1. (a) SEM image and (b) EDS mapping spectrum of commercial MoS_2.

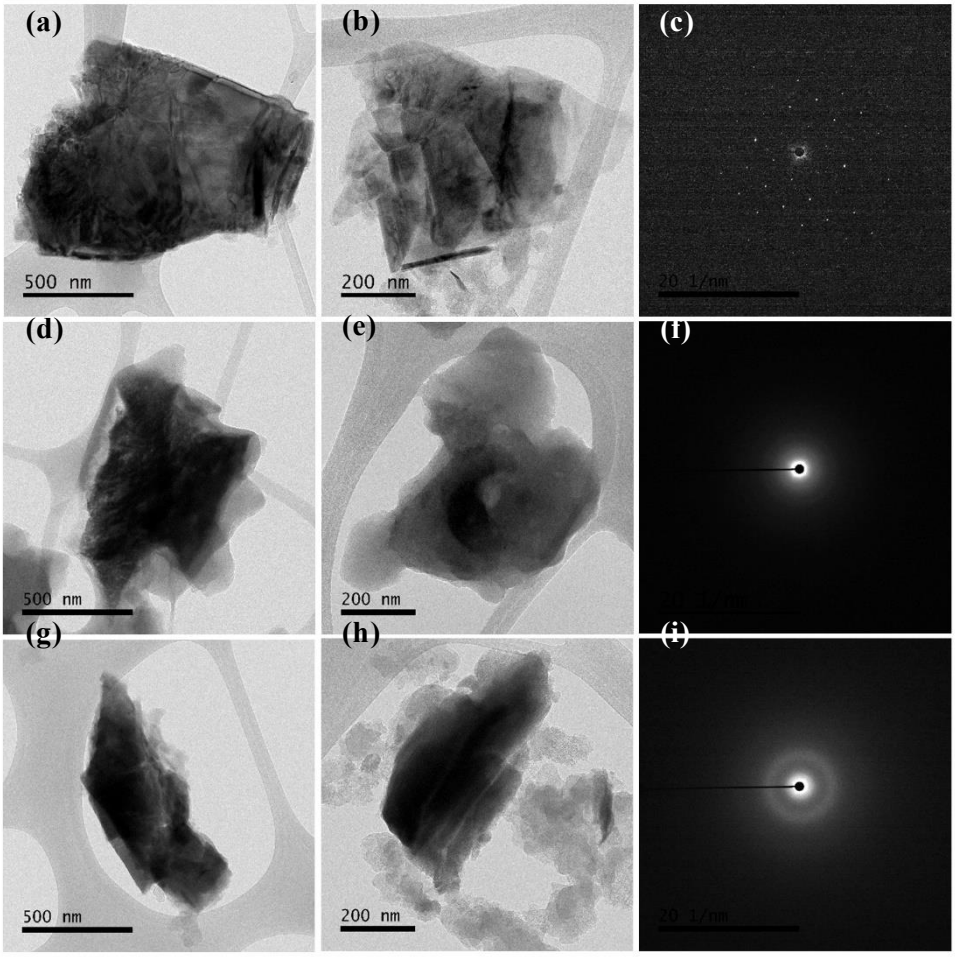
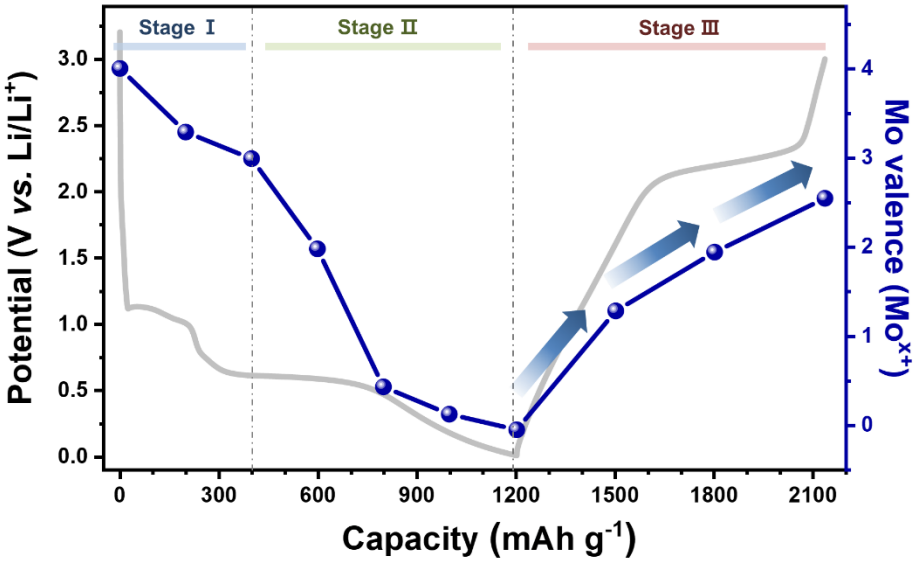


Figure S2. Low-magnitude TEM images of a MoS_2 microsphere for (a,b) pristine, (d,e) fully discharged, and (g,h) fully charged MoS_2. Corresponding ED patterns of the MoS_2 electrode in (c,f,i), respectively.



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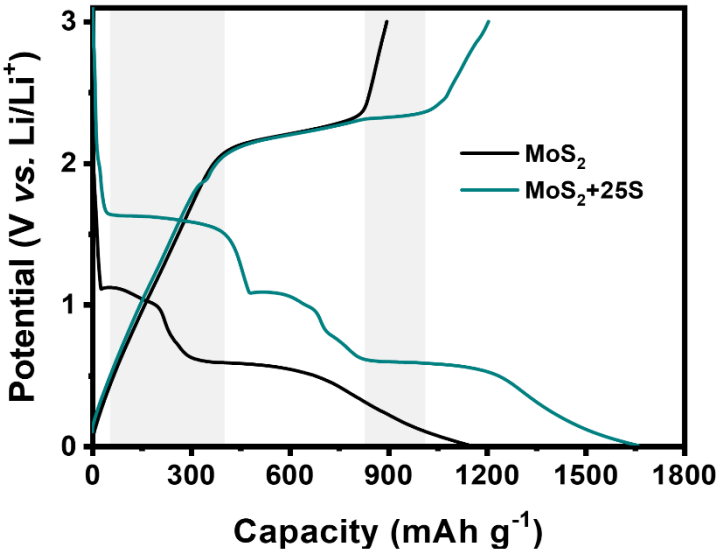

Figure S4. Voltage profiles for MoS_2 and MoS_2+25S composite during the first cycle at 100 mA g^{-1} between 3.0 and 0.001 V (*vs.* Li/Li^+). The highlighted region indicates that the additional part which is only related to the sulfur reaction.

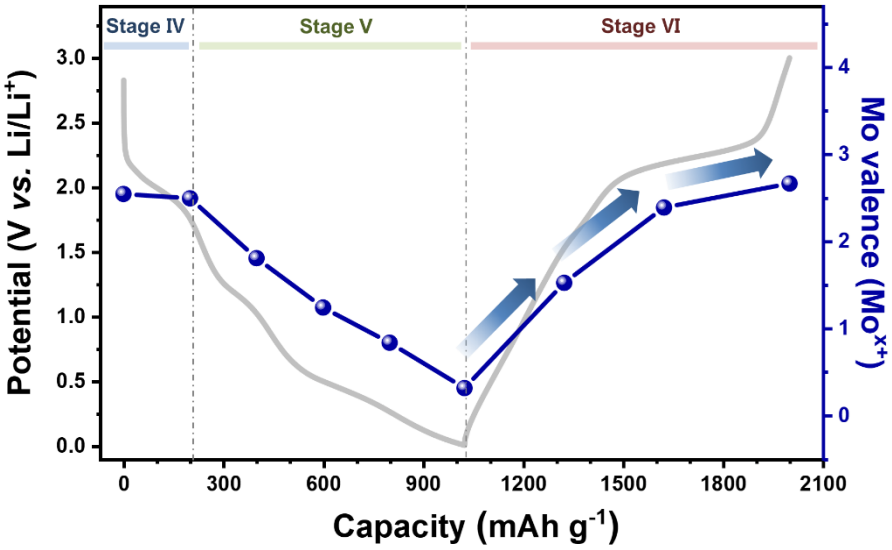

Figure S5. Voltage profile (left, gray line), and Mo valence vs. capacity (right, dark blue line) of MoS_2 electrode upon the second cycle. Mo valence was calculated via linear interpolation using the X-ray absorption edge point of bulk MoS_2, and Mo metal foil as the reference.

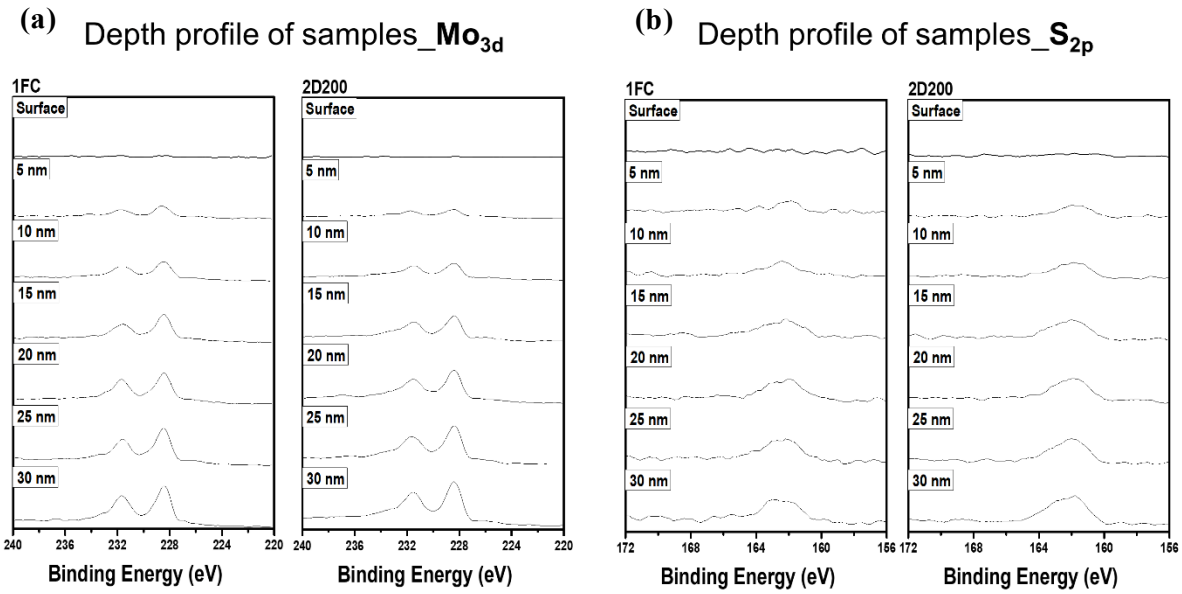

Figure S6. Depth profile XPS of MoS_2, which was obtained before sputtering and after sputtering at 30 nm (5 nm per each level). (a) Mo_{3d} XPS spectra of 1FC and 2D200, (b) S_{2p} XPS spectra of 1FC and 2D200.

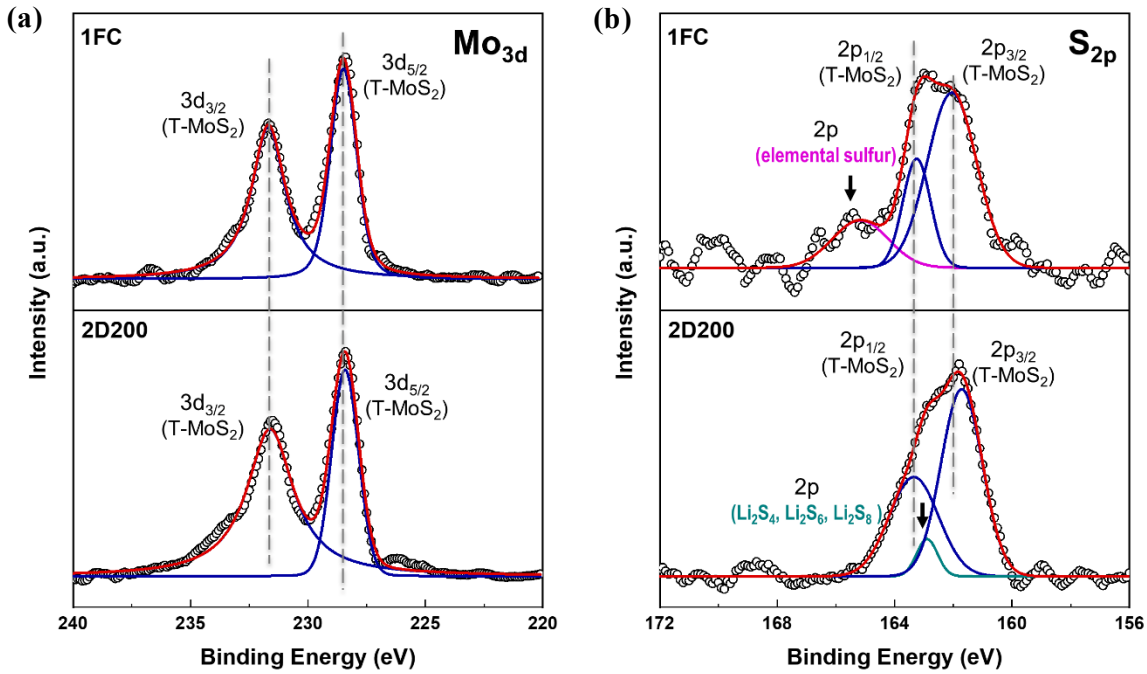

Figure S7. XPS spectra of MoS_2 after eliminating the SEI layer. (a) Mo_{3d} at level 6 (30 nm) and (b) S_{2p} at level 6 (30 nm).

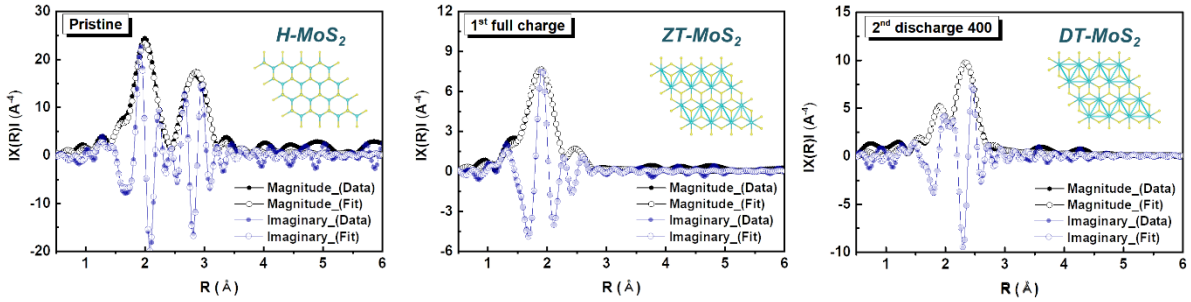

Figure S8. k^3-weighted Fourier transform magnitude and imaginary part along with the best-fit model of Mo K-edge EXAFS spectrum of pristine MoS_2, 1st full charge, and 2nd discharge 400.

Table S1. Best-fit results for k^3-weighted Mo K-edge EXAFS spectrum data.

Sample	Path	CN	R [Å]	σ^2 [10^{-3}Å]	R-factor
Pristine (H-MoS_2)	Mo-S	6.0*	2.407(2)	2.27*	0.00569
	Mo-Mo	6.0*	3.171(3)	4.1(2)	
1FC (ZT-MoS_2)	Mo-S	4.4(2)	2.413(3)	8.0(5)	0.00194
	Mo-Mo	2.1(8)	2.771(10)	16.1(38)	
2D-400 (DT-MoS_2)	Mo-S	3.5(4)	2.475(12)	5.4(10)	0.00209
	Mo-Mo	3.0(4)	2.652(6)	5.0(5)	

*** Fixed value**

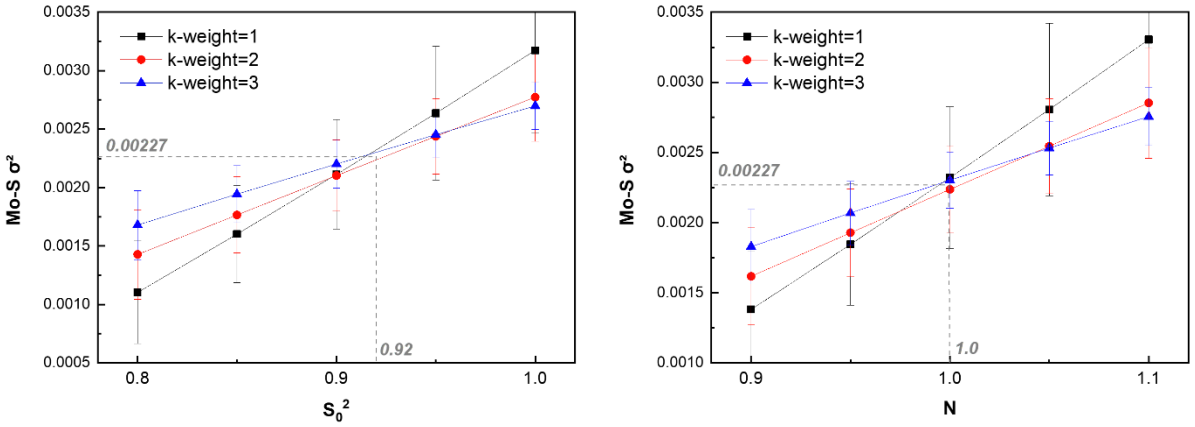

Figure S9. k-weight fitting of pristine MoS_2 EXAFS to determine the reduction factor (S_0^2) and coordination number (CN).

To determine the reduction factor (S_0^2) of MoS_2, k-weight fitting was performed based on the pristine sample as shown in **Figure S9** and the S_0^2 value was obtained as 0.92. EXAFS fitting with the S_0^2 value fixed at 0.92 was performed for two other samples. Consequently, the pristine MoS_2 results were consistent with the H-MoS_2 (hexagonal symmetry) crystallography values.

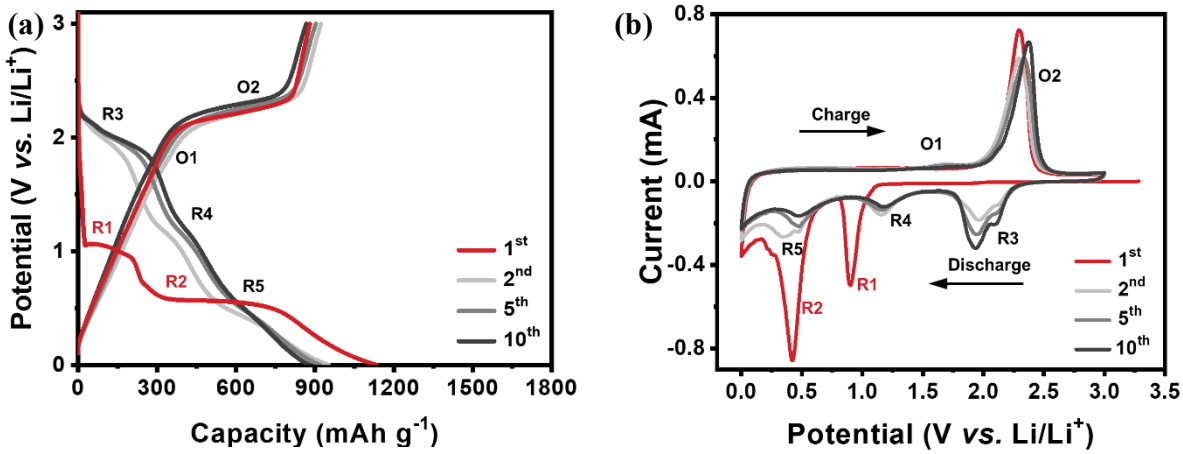

Figure S10. (a) Voltage profiles of the MoS_2 during the 1st to 10th cycles between 0.001–3.0 V (*vs.* Li/Li^+) at a current density of 100 mA g^{-1}. (b) CV profiles between 0.001–3.0 V (*vs.* Li/Li^+) at a scan rate of 0.05 mV s^{-1} during the 1st to 10th cycles.